

PATENT ABSTRACTS OF JAPAN

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(54) PRODUCTION OF POLYETHYLENE TEREPHTHALATE FOAM SHEET

(57)Abstract:

PURPOSE: To continuously produce a beautiful PET foam sheet containing fine air bubbles with a size of 100μm or less, having high mechanical strength and free from surface unevenness.

CONSTITUTION: A process supplying a polyethylene terephthalate resin with intrinsic viscosity of 0.8 or more to an extruder to melt the same and adding a foaming agent based on carbon fluoride having a b.p. of 90–180° C under normal pressure to the molten resin and a process extruding the resin containing the foaming agent to the atmosphere are provided.

LEGAL STATUS

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CLAIMS

[Claim(s)]

[Claim 1] The manufacture approach of the polyethylene terephthalate foaming sheet characterized by providing the process which makes the foaming agent which uses as a principal component the carbon fluoride which has the boiling point of the range of 90-180 degrees C in ordinary pressure contain, and the process which extrudes the resin containing this foaming agent in atmospheric air, and produces a foaming sheet after limiting viscosity supplied and carried out melting of the 0.8 or more polyethylene terephthalate resin to the extruder.

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DETAILED DESCRIPTION

[Detailed Description of the Invention]

[0001]

[Industrial Application] This invention relates to the approach of manufacturing a polyethylene terephthalate foaming sheet suitable as ingredients, such as heat insulating material, a heat insulator, a packing material, packaging, and electronic equipment components.

[0002]

[Description of the Prior Art] Conventionally, as an approach of manufacturing the foaming sheet of polyethylene terephthalate (it abbreviates to PET hereafter), high pressure gas is made to permeate a PET sheet, and the technique (U.S. Pat. No. 4473665) made to foam by heating this more than the softening temperature of resin is known. According to this approach, the foaming object which has detailed air bubbles 100 micrometers or less can be acquired. However, this approach is a batch method and is not suitable for continuation manufacture for obtaining a long picture foaming sheet.

[0003] The technique indicated by JP,2-251543,A is known as an approach of on the other hand manufacturing a PET foaming sheet continuously. After this approach supplies the pyromellitic dianhydride and the sodium carbonate which are PET and a thickener to an extruder, and carries out melting kneading, and making PET thicken and making it into the optimal viscosity for foaming, it is an approach of obtaining a foaming sheet, by pouring in the foaming agent which gasifies or expands with heating from the extruder middle, and extruding under ordinary pressure from a dice. As a foaming agent, inert gas, such as halogenated hydrocarbon, such as hydrocarbons, such as a propane, butane, and a pentane, fluoro carbon 21, and dichloro tetra-FURUOSO ethane, nitrogen gas, and carbon dioxide gas, etc. is used, for example.

[0004] However, when this approach was retested, it became clear that there were the following problems. When foaming agents, such as a hydrocarbon and halogenated hydrocarbon, are used, since the solubility parameter (SP value) of these foaming agents is comparatively close to it of PET, its compatibility with PET is very good. For this reason, although foam is obtained, in order for air bubbles to unite and to grow up to be air bubbles 200 micrometers or more, detailed air bubbles 100 micrometers or less are not obtained. Moreover, when inert gas, such as nitrogen gas and carbon dioxide gas, is used for a foaming agent, it becomes the large foaming sheet of the diameter of air bubbles similarly. And since a gas foaming agent has the quick diffusion rate, in desorbing gas from a sheet and irregularity's arising on a front face, expansion ratio becomes low with 1.5 or less times. Moreover, by this approach, since the thickener is used, coloring of a sheet is accepted.

[0005] In addition, the technique indicated by JP,61-48411,B is known as an approach of manufacturing PET foam continuously. This approach produces string-like continuation foam by the boiling point in ordinary temperature making aromatic polyester melt 8000poise or more contain [melt viscosity / in temperature higher 15 degrees C than the melting point] a foaming agent 90 degrees C or less, and extruding, and heat-treating so that the degree of crystallinity of foam may become 30% or more further. This approach is indicated to have specified the boiling point of a foaming agent as 90 degrees C or less, because good foam is not obtained in order that foam may cause contraction at the time of cooling, if the foaming agent with which the

boiling point exceeds this temperature is used.

[0006] However, even if the boiling point uses a foaming agent 90 degrees C or less, the diameter of air bubbles is imagined to become large by the reason mentioned above compatibility with PET is good. Therefore, also in this approach, it is difficult to obtain continuously the foam in which the diameter of air bubbles has detailed air bubbles 100 micrometers or less.

[0007]

[Problem(s) to be Solved by the Invention] As stated above, by the conventional approach, the diameter of air bubbles has detailed air bubbles 100 micrometers or less, and mechanical strength is large, and it is difficult to obtain continuously a beautiful PET foaming sheet without surface irregularity.

[0008] The diameter of air bubbles has detailed air bubbles 100 micrometers or less, mechanical strength is large and this invention aims at offering the approach that a beautiful PET foaming sheet without surface irregularity can be manufactured continuously.

[0009]

[Means for Solving the Problem] The manufacture approach of the polyethylene terephthalate foaming sheet of this invention is characterized by providing the process which makes the foaming agent which uses as a principal component the carbon fluoride which has the boiling point of the range of 90-180 degrees C in ordinary pressure contain, and the process which extrudes the resin containing this foaming agent in atmospheric air, and produces a foaming sheet, after limiting viscosity supplies and carries out melting of the 0.8 or more polyethylene terephthalate resin to an extruder.

[0010] In this invention, as PET, the high resin of limiting viscosity is desirable, and when it measures with the mixed solvent of a phenol / tetrachloroethane =4/6, what has 0.8 or more values is used. If this condition is fulfilled, addition of the thickener for raising the viscosity of resin which is indicated by JP,2-251543,A etc. is unnecessary. Since a foaming agent ****s more easily than a sheet when limiting viscosity uses resin lower than 0.8, and PET is extruded in atmospheric air from an extruder, it becomes difficult to obtain good foam.

[0011] In addition, various additives, such as a crystallization nucleating additive, a crystallization accelerator, an air-bubbles-ized nucleating additive, an anti-oxidant, an antistatic agent, an ultraviolet-rays inhibitor, a pigment, a color, and lubricant, may be blended in the range which does not spoil the property of resin original.

[0012] In this invention, the solvent which uses as a principal component the carbon fluoride which has the boiling point of the range of 90-180 degrees C in ordinary pressure as a foaming agent is used. Since that SP value is quite smaller than that of PET, the carbon fluoride within the limits of this does not have so good compatibility with PET. For this reason, it is hard coming to unite the air bubbles distributed minutely, and is imagined as what becomes easy to exist with detailed air bubbles. Moreover, also in case a foaming agent gasifies and air-bubbles-izes, the balance of the viscosity of resin and the growth force of air bubbles is good, and the foaming sheet which has detailed air bubbles 100 micrometers or less is obtained.

[0013] Having specified the boiling point of a foaming agent as mentioned above is based on the following reasons. When the foaming agent which has the boiling point higher than 180 degrees C is used, since the difference of the boiling point of a foaming agent and the temperature (about 270-290 degrees C) of melting resin is small, the force of a foaming agent gasifying and growing up to be air bubbles becomes weak. For this reason, expansion ratio becomes a 1.5 or less-time foaming sheet and is not practical. When the foaming agent which, on the other hand, has the boiling point lower than 90 degrees C is used, in order that the growth force of air bubbles may overcome the viscosity of resin, it becomes difficult to obtain the foaming sheet with which air bubbles grow greatly and have detailed air bubbles 100 micrometers or less.

[0014] In this invention, the process which is made to foam by extruding the PET containing a foaming agent in atmospheric air from an extruder, and produces a foaming sheet can use the well-known approach currently enforced conventionally.

[0015]

[Function] In this invention, limiting viscosity uses the solvent which uses as a principal component the 0.8 or more PET and carbon fluoride whose boiling point is 90-180 degrees C as

a foaming agent. Since solubility parameters (SP value) differ greatly mutually, the solvent which uses as a principal component PET [which has such physical properties], and carbon fluoride has low compatibility, and a foaming agent distributes minutely in resin. Moreover, at the time of foaming, the balance of the viscosity of resin and the cellular growth force is good. For this reason, the rate which the air bubbles minutely distributed in early stages of foaming unite into a cellular growth process is low. Therefore, air bubbles are detailed, it has a beautiful front face without irregularity, and mechanical strength can also obtain a large foaming sheet.

[0016]

[Example] Hereafter, the example of this invention is explained.

[0017] The PET (the Unitika make, SA grade, limiting viscosity = 1.0) dried at 140 degrees C with the example 1 moisture-absorption-and-drying machine for 5 hours or more was supplied to the extruder, and melting kneading was carried out. Next, the pressure when evaporating FURORINATO FC-77 (the Sumitomo 3M make, 97 degrees C of boiling points) in a foaming agent inlet as a foaming agent from the extruder middle is 50kg/cm². It poured in so that it might become, and it was made to dissolve and distribute in resin. Then, it rolled round, having extruded under ordinary pressure with T dice, having produced the sheet, and cooling.

[0018] The sheet was produced like the example 1 except having used the uni-pet (Japanese uni-pet make, RT-580CA, limiting viscosity = 1.1) as example 2 PET.

[0019] The sheet was produced like the example 1 except having used the Mitsui pet (the product made of Mitsui pet resin, J005, limiting viscosity = 1.2) as example 3 PET.

[0020] The sheet was produced like the example 1 except having used the Unitika make and NEH grade (limiting viscosity = 0.88) as example 4 PET.

[0021] The sheet was produced like the example 1 except having used AFURUDO E-10 (the Asahi Glass make, 100 degrees C of boiling points) as example 5 foaming agent.

[0022] The sheet was produced like the example 1 except having used FURORINATO FC-43 (the Sumitomo 3M make, 174 degrees C of boiling points) as example 6 foaming agent.

[0023] The sheet was produced like the example 1 except having used AFURUDO E-18 (the Asahi Glass make, 175 degrees C of boiling points) as example 7 foaming agent.

[0024] The sheet was produced like the example 1 except having used Unitika polyester (the Unitika make, MA-2103, limiting viscosity = 0.68) as example of comparison 1 PET. Instead of example of comparison 2 PET, the sheet was produced like the example 1 except having used polypropylene resin (the Mitsubishi Petrochemical make, BC-8BP).

[0025] The sheet was produced like the example 1 except having used chlorofluorocarbon -218 (the Showa Denko make, -37 degrees C of boiling points) as example of comparison 3 foaming agent.

[0026] The sheet was produced like the example 1 except having used FURORINATO FC-70 (the Sumitomo 3M make, 215 degrees C of boiling points) as example of comparison 4 foaming agent.

[0027] The sheet was produced like the example 1 except having used Freon 113 (made in Japanese East FURORO Chemical, 48 degrees C of boiling points) as example of comparison 5 foaming agent.

[0028] The sheet was produced like the example 1 except having used toluene (the Wako Pure Chem make, 115 degrees C of boiling points) as example of comparison 6 foaming agent.

[0029] each obtained sheet — the diameter of air bubbles, expansion ratio, mechanical strength, and a front face — the result of having investigated description is shown in Table 1.

[0030]

[Table 1]

	樹脂種類	極限粘度	発泡剤種類	沸点(°C)	気泡径(μm)	発泡倍率	機械強度	表面凹凸
実施例1	PET (SA)	1. 0	フロリナート FC-77	97 <100	4. 5	大	なし	
実施例2	PET (RJ-580CA)	1. 1	フロリナート FC-77	97 <100	4. 9	大	なし	
実施例3	PET (U055)	1. 2	フロリナート FC-77	97 <100	4. 7	大	なし	
実施例4	PET (NEW)	0. 88	フロリナート FC-77	97 <100	4. 0	大	なし	
実施例5	PET (SA)	1. 0	アフルード E-10	100 <100	3. 9	大	なし	
実施例6	PET (SA)	1. 0	フロリナート FC-43	174 <100	3. 2	大	なし	
実施例7	PET (SA)	1. 0	アフルード E-18	175 <100	3. 4	大	なし	
比較例1	PET (MA-2103)	0. 68	フロリナート FC-77	97 500	1. 9	小	大	
比較例2	PP (BC-BP)		フロリナート FC-77	97 <100	1. 5	中	なし	
比較例3	PET (SA)	1. 0	フロリナート FC-77	-37 300	3. 8	小	中	
比較例4	PET (SA)	1. 0	フロリナート FC-10	215 発泡せず	-	-	-	
比較例5	PET (SA)	1. 0	フレオーン113	48 350	4. 9	小	大	
比較例6	PET (SA)	1. 0	トルエン	115 250	2. 8	小	中	

The diameter of air bubbles had detailed air bubbles 100 micrometers or less, and the PET foaming sheet of examples 1-6 has satisfied all the properties that expansion ratio is high, and mechanical strength is large, and no surface irregularity is so that clearly from Table 1.

[0031]

[Effect of the Invention] If the approach of this invention is used as explained in full detail above, the diameter of air bubbles has detailed air bubbles 100 micrometers or less, mechanical strength is large and a beautiful PET foaming sheet without surface irregularity can be manufactured continuously.

[Translation done.]

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TECHNICAL FIELD

[Industrial Application] This invention relates to the approach of manufacturing a polyethylene terephthalate foaming sheet suitable as ingredients, such as heat insulating material, a heat insulator, a packing material, packaging, and electronic equipment components.

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PRIOR ART

[Description of the Prior Art] Conventionally, as an approach of manufacturing the foaming sheet of polyethylene terephthalate (it abbreviates to PET hereafter), high pressure gas is made to permeate a PET sheet, and the technique (U.S. Pat. No. 4473665) made to foam by heating this more than the softening temperature of resin is known. According to this approach, the foaming object which has detailed air bubbles 100 micrometers or less can be acquired. However, this approach is a batch method and is not suitable for continuation manufacture for obtaining a long picture foaming sheet.

[0003] The technique indicated by JP,2-251543,A is known as an approach of on the other hand manufacturing a PET foaming sheet continuously. After this approach supplies the pyromellitic dianhydride and the sodium carbonate which are PET and a thickener to an extruder, and carries out melting kneading, and making PET thicken and making it into the optimal viscosity for foaming, it is an approach of obtaining a foaming sheet, by pouring in the foaming agent which gasifies or expands with heating from the extruder middle, and extruding under ordinary pressure from a dice. As a foaming agent, inert gas, such as halogenated hydrocarbon, such as hydrocarbons, such as a propane, butane, and a pentane, fluoro carbon 21, and dichloro tetra-FURUOSO ethane, nitrogen gas, and carbon dioxide gas, etc. is used, for example.

[0004] However, when this approach was retested, it became clear that there were the following problems. When foaming agents, such as a hydrocarbon and halogenated hydrocarbon, are used, since the solubility parameter (SP value) of these foaming agents is comparatively close to it of PET, its compatibility with PET is very good. For this reason, although foam is obtained, in order for air bubbles to unite and to grow up to be air bubbles 200 micrometers or more, detailed air bubbles 100 micrometers or less are not obtained. Moreover, when inert gas, such as nitrogen gas and carbon dioxide gas, is used for a foaming agent, it becomes the large foaming sheet of the diameter of air bubbles similarly. And since a gas foaming agent has the quick diffusion rate, in desorbing gas from a sheet and irregularity's arising on a front face, expansion ratio becomes low with 1.5 or less times. Moreover, by this approach, since the thickener is used, coloring of a sheet is accepted.

[0005] In addition, the technique indicated by JP,61-48411,B is known as an approach of manufacturing PET foam continuously. This approach produces string-like continuation foam by the boiling point in ordinary temperature making aromatic polyester melt 8000poise or more contain [melt viscosity / in temperature higher 15 degrees C than the melting point] a foaming agent 90 degrees C or less, and extruding, and heat-treating so that the degree of crystallinity of foam may become 30% or more further. This approach is indicated to have specified the boiling point of a foaming agent as 90 degrees C or less, because good foam is not obtained in order that foam may cause contraction at the time of cooling, if the foaming agent with which the boiling point exceeds this temperature is used.

[0006] However, even if the boiling point uses a foaming agent 90 degrees C or less, the diameter of air bubbles is imagined to become large by the reason mentioned above compatibility with PET is good. Therefore, also in this approach, it is difficult to obtain continuously the foam in which the diameter of air bubbles has detailed air bubbles 100 micrometers or less.

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EFFECT OF THE INVENTION

[Effect of the Invention] If the approach of this invention is used as explained in full detail above, the diameter of air bubbles has detailed air bubbles 100 micrometers or less, mechanical strength is large and a beautiful PET foaming sheet without surface irregularity can be manufactured continuously.

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TECHNICAL PROBLEM

[Problem(s) to be Solved by the Invention] As stated above, by the conventional approach, the diameter of air bubbles has detailed air bubbles 100 micrometers or less, and mechanical strength is large, and it is difficult to obtain continuously a beautiful PET foaming sheet without surface irregularity.

[0008] The diameter of air bubbles has detailed air bubbles 100 micrometers or less, mechanical strength is large and this invention aims at offering the approach that a beautiful PET foaming sheet without surface irregularity can be manufactured continuously.

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MEANS

[Means for Solving the Problem] The manufacture approach of the polyethylene terephthalate foaming sheet of this invention is characterized by providing the process which makes the foaming agent which uses as a principal component the carbon fluoride which has the boiling point of the range of 90-180 degrees C in ordinary pressure contain, and the process which extrudes the resin containing this foaming agent in atmospheric air, and produces a foaming sheet, after limiting viscosity supplies and carries out melting of the 0.8 or more polyethylene terephthalate resin to an extruder.

[0010] In this invention, as PET, the high resin of limiting viscosity is desirable, and when it measures with the mixed solvent of a phenol / tetrachloroethane =4/6, what has 0.8 or more values is used. If this condition is fulfilled, addition of the thickener for raising the viscosity of resin which is indicated by JP,2-251543,A etc. is unnecessary. Since a foaming agent **** more easily than a sheet when limiting viscosity uses resin lower than 0.8, and PET is extruded in atmospheric air from an extruder, it becomes difficult to obtain good foam.

[0011] In addition, various additives, such as a crystallization nucleating additive, a crystallization accelerator, an air-bubbles-ized nucleating additive, an anti-oxidant, an antistatic agent, an ultraviolet-rays inhibitor, a pigment, a color, and lubricant, may be blended in the range which does not spoil the property of resin original.

[0012] In this invention, the solvent which uses as a principal component the carbon fluoride which has the boiling point of the range of 90-180 degrees C in ordinary pressure as a foaming agent is used. Since that SP value is quite smaller than that of PET, the carbon fluoride within the limits of this does not have so good compatibility with PET. For this reason, it is hard coming to unite the air bubbles distributed minutely, and is imagined as what becomes easy to exist with detailed air bubbles. Moreover, also in case a foaming agent gasifies and air-bubbles-izes, the balance of the viscosity of resin and the growth force of air bubbles is good, and the foaming sheet which has detailed air bubbles 100 micrometers or less is obtained.

[0013] Having specified the boiling point of a foaming agent as mentioned above is based on the following reasons. When the foaming agent which has the boiling point higher than 180 degrees C is used, since the difference of the boiling point of a foaming agent and the temperature (about 270-290 degrees C) of melting resin is small, the force of a foaming agent gasifying and growing up to be air bubbles becomes weak. For this reason, expansion ratio becomes a 1.5 or less-time foaming sheet and is not practical. When the foaming agent which, on the other hand, has the boiling point lower than 90 degrees C is used, in order that the growth force of air bubbles may overcome the viscosity of resin, it becomes difficult to obtain the foaming sheet with which air bubbles grow greatly and have detailed air bubbles 100 micrometers or less.

[0014] In this invention, the process which is made to foam by extruding the PET containing a foaming agent in atmospheric air from an extruder, and produces a foaming sheet can use the well-known approach currently enforced conventionally.

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OPERATION

[Function] In this invention, limiting viscosity uses the solvent which uses as a principal component the 0.8 or more PET and carbon fluoride whose boiling point is 90–180 degrees C as a foaming agent. Since solubility parameters (SP value) differ greatly mutually, the solvent which uses as a principal component PET [which has such physical properties], and carbon fluoride has low compatibility, and a foaming agent distributes minutely in resin. Moreover, at the time of foaming, the balance of the viscosity of resin and the cellular growth force is good. For this reason, the rate which the air bubbles minutely distributed in early stages of foaming unite into a cellular growth process is low. Therefore, air bubbles are detailed, it has a beautiful front face without irregularity, and mechanical strength can also obtain a large foaming sheet.

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EXAMPLE

[Example]-Hereafter, the example of this invention is explained.

[0017] The PET (the Unitika make, SA grade, limiting viscosity = 1.0) dried at 140 degrees C with the example 1 moisture-absorption-and-drying machine for 5 hours or more was supplied to the extruder, and melting kneading was carried out. Next, the pressure when evaporating FURORINATO FC-77 (the Sumitomo 3M make, 97 degrees C of boiling points) in a foaming agent inlet as a foaming agent from the extruder middle is 50kg/cm². It poured in so that it might become, and it was made to dissolve and distribute in resin. Then, it rolled round, having extruded under ordinary pressure with T dice, having produced the sheet, and cooling.

[0018] The sheet was produced like the example 1 except having used the uni-pet (Japanese uni-pet make, RT-580CA, limiting viscosity = 1.1) as example 2 PET.

[0019] The sheet was produced like the example 1 except having used the Mitsui pet (the product made of Mitsui pet resin, J005, limiting viscosity = 1.2) as example 3 PET.

[0020] The sheet was produced like the example 1 except having used the Unitika make and NEH grade (limiting viscosity = 0.88) as example 4 PET.

[0021] The sheet was produced like the example 1 except having used AFURUDO E-10 (the Asahi Glass make, 100 degrees C of boiling points) as example 5 foaming agent.

[0022] The sheet was produced like the example 1 except having used FURORINATO FC-43 (the Sumitomo 3M make, 174 degrees C of boiling points) as example 6 foaming agent.

[0023] The sheet was produced like the example 1 except having used AFURUDO E-18 (the Asahi Glass make, 175 degrees C of boiling points) as example 7 foaming agent.

[0024] The sheet was produced like the example 1 except having used Unitika polyester (the Unitika make, MA-2103, limiting viscosity = 0.68) as example of comparison 1 PET.

Instead of example of comparison 2 PET, the sheet was produced like the example 1 except having used polypropylene resin (the Mitsubishi Petrochemical make, BC-8BP).

[0025] The sheet was produced like the example 1 except having used chlorofluocarbon -218 (the Showa Denko make, -37 degrees C of boiling points) as example of comparison 3 foaming agent.

[0026] The sheet was produced like the example 1 except having used FURORINATO FC-70 (the Sumitomo 3M make, 215 degrees C of boiling points) as example of comparison 4 foaming agent.

[0027] The sheet was produced like the example 1 except having used Freon 113 (made in Japanese East FURORO Chemical, 48 degrees C of boiling points) as example of comparison 5 foaming agent.

[0028] The sheet was produced like the example 1 except having used toluene (the Wako Pure Chem make, 115 degrees C of boiling points) as example of comparison 6 foaming agent.

[0029] each obtained sheet — the diameter of air bubbles, expansion ratio, mechanical strength, and a front face — the result of having investigated description is shown in Table 1.

[0030]

[Table 1]

	樹種	樹脂 種類	極限粘度	泡種類	沸點 (°C)	気泡径 (μm)	発泡倍率	機械強度	表面凹凸
実施例1	PET (SA)	1. 0	フロリナート PC-77	97	<100	4. 5	大	なし	
実施例2	PET (RT-5800A)	1. 1	フロリナート PC-77	97	<100	4. 9	大	なし	
実施例3	PET (U055)	1. 2	フロリナート PC-77	97	<100	4. 7	大	なし	
実施例4	PET (NEH)	0. 88	フロリナート PC-77	97	<100	4. 0	大	なし	
実施例5	PET (SA)	1. 0	アフルード E-10	100	<100	3. 9	大	なし	
実施例6	PET (SA)	1. 0	フロリナート PC-43	174	<100	3. 2	大	なし	
実施例7	PET (SA)	1. 0	アフルード E-18	175	<100	3. 4	大	なし	
比較例1	PET (WA-2103)	0. 68	フロリナート PC-77	97	500	1. 9	小	大	
比較例2	PP (PC-88P)		フロリナート PC-77	97	<100	1. 5	中	なし	
比較例3	PET (SA)	1. 0	フロン218	-37	300	3. 8	小	中	
比較例4	PET (SA)	1. 0	フロリナート PC-70	215	発泡せず	-	-	-	
比較例5	PET (SA)	1. 0	フレオジン113	48	350	4. 9	小	大	
比較例6	PET (SA)	1. 0	トルエン	115	250	2. 8	小	中	

The diameter of air bubbles had detailed air bubbles 100 micrometers or less, and the PET foaming sheet of examples 1-6 has satisfied all the properties that expansion ratio is high, and mechanical strength is large, and no surface irregularity is so that clearly from Table 1.
[0031]

[Translation done.]